

(19)



Europäisches Patentamt
European Patent Office
Office européen des brevets



(11) Publication number:

0 291 074 B1

(12)

EUROPEAN PATENT SPECIFICATION(45) Date of publication of patent specification: **29.01.92** (51) Int. Cl.⁵: **C07F 9/54**(21) Application number: **88107735.8**(22) Date of filing: **13.05.88**(54) **Process for producing quaternary salts.**

(30) Priority: **13.05.87 JP 116463/87**
14.05.87 JP 117380/87
22.05.87 JP 125427/87
22.05.87 JP 125428/87

(43) Date of publication of application:
17.11.88 Bulletin 88/46

(45) Publication of the grant of the patent:
29.01.92 Bulletin 92/05

(84) Designated Contracting States:
DE FR GB NL

(56) References cited:
EP-A- 0 269 949
US-A- 4 266 079

DE-A-B24673IVc/129, 1955 (B.A.S.F)

CHEMICAL ABSTRACTS, vol.108, no.22,
30.05.88, page 535, column 2, abstract
194813j, Columbus, Ohio, USA; & JP-
A-63-24080, 01.02.88 (Cat. A,P)

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EP 0 291 074 B1

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DescriptionFIELD OF THE INVENTION

5 This invention relates to a process for producing quaternary salts of high purity efficiently. More particularly, the invention relates to a process for producing a quaternary phosphonium salt by reacting a tertiary phosphine with a carbonic acid diester to form a corresponding quaternary carbonate and further mixing it with an acid to perform decarboxylation.

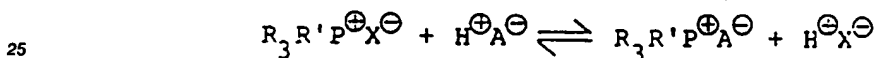
The quaternary salts obtained by the process of this invention are useful compounds which are used in
10 wide fields as various catalysts such as a phase transfer catalyst, etc., electrolytes for aqueous or organic electrolytic solutions, various additives, medicaments, etc.

BACKGROUND OF THE INVENTION

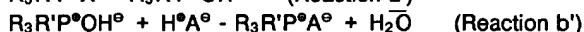
15 As a process of synthesizing a quaternary phosphonium salt, quaternarizing a tertiary phosphine with an alkyl halide, a dialkylsulfuric acid, etc., under heating as shown in the equation:



20 and then performing, in equilibrium, an anion exchange by the reaction shown in the following equation is generally known:



Also, as a process for producing a high pure quaternary phosphonium salt, a process of once converting a quaternary phosphonium halide into a quaternary phosphonium hydroxide (Reaction a') and
30 then neutralizing the hydroxide with an acid corresponding to the anion of the desired salt (Reaction b') as shown in the following equations is most general:



35 Furthermore, the processes for producing the quaternary phosphonium hydroxide by aforesaid Reaction a' are expensive as an industrial process same as in the aforesaid cases of producing a quaternary ammonium hydroxide and, in particular, in the case of producing a high-pure product wherein the halide content in the quaternary phosphonium hydroxide is controlled to an order of ppm, the production cost also becomes considerably high.

DE-A-B24673IVc/12q discloses a method for preparing quaternary ammonium salts of carbonic acid by reacting a tertiary amine with an ester of a carbonic acid. A preparative method is disclosed in which a tertiary amine is heated with dimethyl carbonate. The quaternary ammonium salt formed in this reaction is then reacted in turn with tartaric acid to form the corresponding quaternary ammonium tartarate salt.

45 US-A-4 266 079 discloses the preparation of a tetrahydrocarbylphosphonium bicarbonate salt from a trihydrocarbyl(hydrocarbylcarboxymethyl) phosphonium hydroxide inner salt.

SUMMARY OF THE INVENTION

50 An object of this invention is to provide a novel process capable of producing a high-pure quaternary salt efficiently as compared with conventional processes of once producing a quaternary phosphonium hydroxide.

Other object of this invention is to provide a novel technique capable of producing a high-pure quaternary salt even in the case that the desired product is a salt of a relatively weak acid such as an
55 organic acid.

Thus, according to this invention, there is provided a process for producing a quaternary salt comprising

(a) a 1st step of reacting a phosphine with a carbonic acid diester to produce a corresponding quaternary

carbonate, and

(b) a 2nd step of mixing the quaternary carbonate thus formed with an acid, while removing carbon dioxide generated from the system, to anion-exchange the carbonate into the corresponding acid.

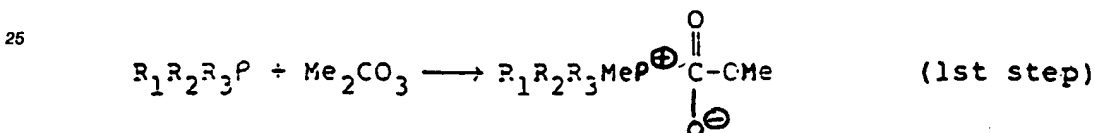
5 DETAILED DESCRIPTION OF THE INVENTION

As the tertiary phosphine which is a raw material in this invention, there are saturated aliphatic phosphines such as trimethylphosphine, diethylmethylphosphine, triethylphosphine, tri-n-propylphosphine, tri-n-butylphosphine, tri-n-pentylphosphine, tri-i-butylphosphine, di-n-butylmethylphosphine, tricyclohexylphosphine, 1,2-bis(dimethylphosphine)ethane, etc.; unsaturated aliphatic phosphines such as triarylphosphine, etc.; aromatic phosphines such as triphenylphosphine, tribenzylphosphine, di-n-propylphosphine, diethylphenylphosphine, n-butyl-diphenylphosphine, etc.; and cyclic phosphines such as 1-ethylphosphoran, 1-phenylphosphoran, 1-phenylphosphane, 1-phenylphosphepane, etc.

As the carbonic acid diester, there are dimethyl carbonate, ethylmethyl carbonate, diethyl carbonate, dipropyl carbonate, etc., but a carbonic acid diester having an alkyl moiety of small carbon atom number such as dimethyl carbonate is preferred since, in this case, the quaternarization reaction proceeds fast.

The quaternarization reaction which is the 1st step in this invention is performed using a tertiary phosphine and a carbonic acid diester in a mol ratio of from 0.2 to 5, and preferably from 0.3 to 3, in the presence or absence of a solvent, at reaction temperature of from 20°C to 200°C, and preferably from 30°C to 160°C.

When a tertiary phosphine and dimethyl carbonate are used as the raw materials, the reaction occurring is shown by the following equation:

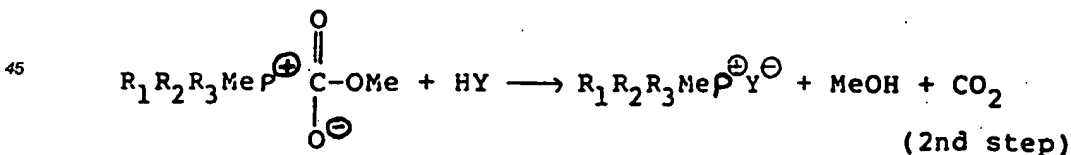


wherein $R_1R_2R_3$ represents a hydrocarbon residue of the tertiary phosphine.

Usually, when the tertiary phosphine is sufficiently converted into a quaternarized product, distilling off the unreacted phosphine and the unreacted carbonic acid diester together with the solvent, in the case of using a solvent, is performed, and the product, if necessary, recrystallized from a suitable organic solvent is then sent to the 2nd step.

In the 2nd step, a stoichiometric amount or a little excessive amount of an acid having the anion of the desired salt as a conjugated base is usually added dropwise to the quaternary carbonate in the presence or absence of a solvent and carbon dioxide generated is removed from the system under reduced pressure or by blowing an inert gas into the reaction system.

In the case of using quaternary phosphonium methylcarbonate, the reaction occurring is shown by the following equation:



wherein Y represents a conjugated base of the acid being used.

There is no particular restriction on the acid being used in the 2nd step, but a stronger acid as compared to carbonic acid completes faster the anion exchange. However, even in the case of using an acid similar to carbonic acid or weaker than carbonic acid, the anion exchange can be carried out by removing the carbonate as carbon dioxide in the system to shift the equilibrium.

Specific examples of the inorganic acid are HF, HCl, HBr, HI, HNO₃, H₂SO₄, H₃PO₃, H₃BO₃, HClO₄, HBF₄, HPF₆, HSbF₆, HAsF₆, HOSO₂Cl, HOSO₂F, H₂CrO₄, H₂S₂O₆, HMnO₄, HReO₄, H₂SeO₄, HSCN, etc.

In the quaternary salts obtained, the salts having EF_4^- , PF_6^- , ClO_4^- , etc., as the conjugated base are

preferred.

In this invention, a desired high-pure quaternary salt is obtained even in the case of using a relatively weak acid such as an organic acid, which makes the process of this invention advantageous.

Specific examples of the organic acid are aliphatic monocarboxylic acids such as formic acid, acetic acid, propionic acid, butyric acid, valeric acid, caproic acid, enanthic acid, caprylic acid, pelargonic acid, capric acid, undecanic acid, lauric acid, tridecanic acid, myristic acid, pentacanic acid, palmitic acid, heptadecanic acid, stearic acid, nonadecanic acid, arachidic acid, isobutyric acid, isovaleric acid, isocaproic acid, ethylbutyric acid, methylvaleric acid, isocaprylic acid, propylvaleric acid, ethylcaproic acid, isocapric acid, tuberculostearic acid, pivalic acid, 2,2-dimethylbutanic acid, 2,2-dimethylpentanic acid, 2,2-dimethylhexanic acid, 2,2-dimethylheptanic acid, 2,2-dimethyloctanic acid, 2-methyl-2-ethylbutanic acid, 2-methyl-2-ethylpentanic acid, 2-methyl-2-ethylhexanic acid, 2-methyl-2-ethylheptanic acid, 2-methyl-2-ethyl-octanic acid, 2-methyl-2-propylpentanic acid, 2-methyl-2-propylhexanic acid, 2-methyl-2-propylheptanic acid, acrylic acid, crotonic acid, isocrotonic acid, 3-butenic acid, pentenic acid, hexenic acid, heptenic acid, octenic acid, nonenic acid, decenic acid, undecenic acid, dodecenic acid, tuzuic acid, physteric acid, goshuyic acid, palmitoleic acid, petroselinic acid, oleic acid, elaidic acid, vaccenic acid, gadoleic acid, methacrylic acid, 3-methylcrotonic acid, tiglic acid, methylpentenic acid, cyclopentacarboxylic acid, cyclohexanecarboxylic acid, trifluoroacetic acid, phenylacetic acid, chloroacetic acid, glycolic acid, lactic acid, etc.; aliphatic polycarboxylic acids such as citric acid, malonic acid, succinic acid, glutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid, undecane diacid, dodecane diacid, tridecane diacid, tetradecane diacid, pentadecane diacid, hexadecane diacid, heptadecane diacid, octadecane diacid, nonadecane diacid, eicosane diacid, methylmalonic acid, ethylmalonic acid, propylmalonic acid, butylmalonic acid, pentylmalonic acid, hexylmalonic acid, dimethylmalonic acid, methylethylmalonic acid, diethylmalonic acid, methylpropylmalonic acid, methylbutylmalonic acid, ethylpropylmalonic acid, dipropylmalonic acid, ethylbutylmalonic acid, propylbutylmalonic acid, dibutylmalonic acid, methylsuccinic acid, ethylsuccinic acid, 2,2-dimethylsuccinic acid, 2,3-dimethylsuccinic acid, 2-methylglutaric acid, maleic acid, citraconic acid, itaconic acid, methyleneglutaric acid, monomethyl maleate, 1,5-octanedicarboxylic acid, 5,6-decanedicarboxylic acid, 1,7-decanedicarboxylic acid, 4,6-dimethyl-4-nonene-1,2-dicarboxylic acid, 4,6-dimethyl-1,2-nonanedicarboxylic acid, 1,7-dodecanedicarboxylic acid, 5-ethyl-1,10-decanedicarboxylic acid, 6-methyl-6-dodecene-1,12-dicarboxylic acid, 6-methyl-1,12-dodecanedicarboxylic acid, 6-ethylene-1,12-dodecanedicarboxylic acid, 7-methyl-7-tetradecene-1,14-dicarboxylic acid, 7-methyl-1,14-tetradecanedicarboxylic acid, 3-hexyl-4-decene-1,2-dicarboxylic acid, 3-hexyl-1,12-decanedicarboxylic acid, 6-ethylene-9-hexadecene-1,16-dicarboxylic acid, 6-ethyl-1,16-hexadecanedicarboxylic acid, 6-phenyl-1,12-dodecanedicarboxylic acid, 7,12-dimethyl-7,11-octadecanediene-1,18-dicarboxylic acid, 7,12-dimethyl-1,18-octadecanedicarboxylic acid, 6,8-diphenyl-1,14-tetradecanedicarboxylic acid, 1,1-cyclopentanedicarboxylic acid, 1,1-cyclopentanedicarboxylic acid, 1,2-cyclopentanedicarboxylic acid, 1,1-cyclohexanedicarboxylic acid, 1,2-cyclohexanedicarboxylic acid, 4-cyclohexene-1,2-dicarboxylic acid, 5-norbornene-2,3-dicarboxylic acid, malic acid, glutamic acid, tartaric acid, citric acid, etc.; aromatic monocarboxylic acids (including o-, m-, and p-isomers) such as benzoic acid, toluic acid, ethylbenzoic acid, propylbenzoic acid, isopropylbenzoic acid, butylbenzoic acid, sec-butylbenzoic acid, tert-butylbenzoic acid, hydroxybenzoic acid, anisic acid, ethoxybenzoic acid, propoxybenzoic acid, isopropoxybenzoic acid, butoxybenzoic acid, isobutoxybenzoic acid, sec-butoxybenzoic acid, tert-butoxybenzoic acid, aminobenzoic acid, N-methylaminobenzoic acid, N-ethylaminobenzoic acid, N-propylaminobenzoic acid, N-isopropylaminobenzoic acid, N-butylaminobenzoic acid, N-isobutylaminobenzoic acid, N-sec-butylaminobenzoic acid, N-tert-butylaminobenzoic acid, N,N-dimethylaminobenzoic acid, N,N-diethylaminobenzoic acid, nitrobenzoic acid, fluorobenzoic acid, resorcinic acid, etc.; aromatic polycarboxylic acids such as phthalic acid, isophthalic acid, terephthalic acid, nitrophthalic acid, trimellitic acid, hemimellitic acid, trimesic acid, pyromellitic acid, etc., and phenols such as phenol, p-fluorophenol, β -naphthol, o-nitrophenol, p-nitrophenol, paminophenol, catechol, resorcin, 2-chlorophenol, 2,4-dichlorophenol, 4,4'-dihydroxydiphenyl-2,2-propane, etc.

In the quaternary salts obtained, those having the anion of a dicarboxylic acid such as maleic acid, phthalic acid, etc., as the conjugated base are preferred.

After the reaction is over, a by-produced alcohol and, if a solvent is used, the solvent are distilled off to provide a desired quaternary salt as solids. If necessary, by recrystallizing the product from a suitable solvent, a high-pure product can be obtained.

Also, when a little excessive amount of an acid to the stoichiometric amount is used for completely removing the carbonic acid ions, the excess acid can be removed by a treatment such as recrystallization, etc.

In the case of producing a quaternary salt of a carboxylic acid in the process of this invention, a carboxylic acid anhydride can be used as the acid in the 2nd step. In this case, it sometimes happens that

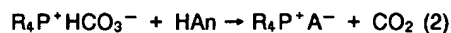
the quaternary salt having a sufficient purity is not obtained, since a carboxylic acid alkyl ester is formed.

Accordingly, by dissolving the quaternary methyl carbonate obtained in the 1st step using, dimethyl carbonate as the carbonic acid diester in water, after removing methanol formed from the system, and mixing the solution with a carboxylic acid anhydride, while removing carbon dioxide generated from the system, a corresponding carboxylate can be produced at high purity without forming the methyl ester.

There is no particular restriction on the carboxylic acid anhydride but specific examples thereof are aliphatic monocarboxylic acid anhydride such as acetic anhydride, propionic anhydride, etc.; aliphatic dicarboxylic acid anhydrides such as maleic anhydride, citraconic anhydride, 1,2-cyclohexanedicarboxylic anhydride, etc.; and aromatic polycarboxylic acid anhydrides such as phthalic anhydride, nitrophthalic anhydride (including 3-nitro compound and 4-nitro compound), trimellitic anhydride, pyromellitic anhydride, etc.

As the quaternary salts obtained, those from a dibasic acid anhydride such as maleic anhydride, phthalic anhydride, etc., are preferred.

In the case of producing a quaternary phosphonium salt, the reaction occurring in this case is shown by the following equation:



wherein HAn represents a carboxylic acid anhydride and A represents a conjugated base thereof.

In the process, the quaternary methyl carbonate is first dissolved in water and after shifting the equilibrium of equation (1) to the right side by removing MeOH formed from the system, the solution is mixed with a carboxylic acid anhydride to cause the reaction as shown in equation (2).

In this case, the step of removing MeOH from the reaction system is performed in the existence of water at reaction temperature of from 0°C to 200°C, preferably from 20°C to 80°C under reduced pressure or normal pressure.

The amount of water is usually from 0.1 to 100, and preferably from 0.5 to 10 by weight based on the amount of the quaternary methyl carbonate.

It is desirable to remove MeOH to an extent that the content of MeOH becomes less than 11%, and preferably less than 6.0% to the amount of the quaternary bicarbonate.

Also, in the reaction step with a carboxylic acid anhydride, a stoichiometric amount or a little excess amount of the carboxylic acid anhydride is mixed with a quaternary bicarbonate in the existence of water and carbon dioxide generated at reaction temperature of from 0°C to 200°C, and preferably from 20°C to 80°C is removed from the system under reduced pressure or normal pressure.

The mixing of a carboxylic acid anhydride is performed by adding dropwise an aqueous solution of a quaternary bicarbonate (95 wt% to 0.1 wt%, and preferably from 70 to 10 wt%) to a suspension of the carboxylic acid anhydride in water of from 0.5 to 100, and preferably 1 to 10 by weight ratio to the anhydride, or adding the carboxylic acid anhydride alone or a mixture of the carboxylic acid anhydride and water of from 0.01 to 100 by weight ratio to an aqueous solution of a quaternary bicarbonate (95 to 0.1 wt%, and preferably 70 to 10 wt%).

After the reaction is over, the solvent is distilled off to provide the desired quaternary carboxylate as solids. If necessary, by recrystallizing the product from a suitable solvent, a high-pure product can be obtained.

As described above, according to the process of this invention, various kinds of quaternary phosphonium salts can be efficiently produced and also even in the case of using a relatively weak acid such as an organic acid, the desired products can be obtained at high purity, which is the large feature of this invention.

Then, the invention is explained in more practically by the following examples.

EXAMPLE 1

(1st Step)

In a stirring system autoclave were filled 9.0 g of dimethyl carbonate and 18.5 g of tri-n-butylphosphine and they were reacted for 15 hours at reaction temperature of 115 °C and reaction pressure of 5.0 kg/cm². After the reaction was over, the autoclave was cooled and then the reaction mixture was collected and analyzed by gas chromatography. The conversion of the tri-n-butylphosphine was 72.8% and the amount of the solid product after distilling off the unreacted materials and the solvent was 17.5 g (63.6% of the theoretical yield). From the elemental analysis and ¹H-NMR, the solid product was confirmed to be tri-n-butylmethylphosphonium carbonate.

(2nd Step)

In 10.0 g of water was dissolved 10.0 g of tri-n-butylmethylphosphonium carbonate and 7.9 g of an aqueous solution of 42% HBF₄ was gradually added to the solution, whereby carbon dioxide simultaneously generated vigorously. For more completely removing carbon dioxide from the system, degassing was performed for 2 hours at 40 °C and 20 mmHg and after confirming carbon dioxide being less than 20 ppm, water was distilled off. The residue formed was recrystallized from a mixture of water and methanol to provide 10.1 g (92.4% of the theoretical yield, yield of 58.8 % to tri-n-butylphosphine) of tri-n-butylmethylphosphonium tetrafluoroborate.

EXAMPLE 2

(1st Step)

By following the same procedure as the 1st step of Example 1 except that 9.0 g of dimethyl carbonate, 11.8 g of triethylphosphine, and 10.0 g of methanol as solvent were used, 17.2 g (78.9% of the theoretical yield) of a solid product was obtained. From the elemental analysis and ¹H-NMR, the solid product was confirmed to be triethylmethylphosphonium carbonate.

(2nd Step)

By following the same procedure as the 2nd step of Example 1 except that 10.0 g of triethylmethylphosphonium carbonate, 11.3 g of an aqueous solution of 42% HBF₄, and 10.0 g of water, 10.8 g (95.3% of the theoretical yield, yield of 79.2% to triethylphosphine) of triethylmethylphosphonium tetrafluoroborate was obtained.

EXAMPLE 3

(1st Step)

By following the same procedure as the 1st step of Example 1 except that 9.0 g of dimethyl carbonate, 17.8 g of 1-phenylphosphane, and 10.0 g of methanol as solvent were used, 19.1 g (71.3% of the theoretical yield) of a solid product was obtained. From the elemental analysis and ¹H-NMR, the solid product was confirmed to be 1-methylphenylphosphonium carbonate.

(2nd Step)

By following the same procedure as the 2nd step of Example 1 - except that 10.0 g of 1-methylphenylphosphonium carbonate, 6.9 g of 60% perchlorate, and 10.0 g of water were used, 10.9 g (94.8% of the theoretical yield, yield of 67.6% to 1-phenylphosphane) of methylphosphonium perchlorate was obtained.

EXAMPLE 4

(1st Step)

By following the same procedure as the 1st step of Example 1 except that 9.0 g of dimethyl carbonate, 21.2 g of triphenylphosphine, and 10.0 g of methanol as solvent were used, 21.3 g (60.5% of the theoretical yield) of a solid product was obtained. From the elemental analysis and ¹H-NMR, the solid product was confirmed to be triphenylmethylphosphonium carbonate.

(2nd Step)

By following the same procedure as the 2nd step of Example 1 except that 10.0 g of triphenylmethylphosphonium carbonate, 6.2 g of 60% prechlorate, and 100 g of ethanol as solvent were used, 10.3 g (92.8% of the theoretical yield, yield of 56.1% to triphenylphosphine) of triphenylmethylphosphonium perchlorate was obtained.

EXAMPLE 5

In 10.0 g of water was dissolved 10.0 g of tri-n-butylmethylphosphonium carbonate obtained as in Example 1 (1st step) and a solution of 4.2 g of maleic acid dissolved in 5.0 g of water was gradually added to the solution, whereby carbon dioxide simultaneously generated vigorously. For more completely removing carbon dioxide, degassing was performed for 2 hours at 40 °C and 20 mmHg and after confirming carbonate ions being less than 2 ppm, water was distilled off. The residue formed was recrystallized from methyl ethyl ketone to produce 11.2 g (94.1% of the theoretical yield, yield of 59.8% to tri-n-butylphosphine) of tri-n-butylmethylphosphonium monomaleate.

EXAMPLE 6

By following the same procedure as Example 5 except that 10.0 g of triethylmethylphosphonium carbonate obtained in Example 1 (1st step), 7.5 g of adipic acid, and 10.0 g of water, 13.5 g (94.4% of the theoretical yield, yield of 74.5% to triethylphosphine) of triethylmethylphosphonium mono adipate was obtained.

EXAMPLE 7

By following the same procedure as Example 5 except that 10.0 g 1-methylphenylphosphonium carbonate obtained as in the 1st step of Example 3, 3.7 g of phenol, and 10.0 g of water were used, 10.2 g (90.3% of the theoretical yield, yield of 64.4% to 1-phenylphosphane) of 1-methylphenylphosphonium phenolate was obtained.

EXAMPLE 8

By following the same procedure as Example 5 except that 10.0 g of triphenylmethylphosphonium carbonate obtained as in the 1st step of Example 4, 1.8 g of acetic acid, and 10.0 g of water, 9.6 g (96.6% of the theoretical yield, yield of 58.4% to triphenylphosphine) of triphenylmethylphosphonium acetate was obtained.

Claims

1. A process for producing a quaternary salt comprising
 - (a) a 1st step of reacting a phosphine with a carbonic acid diester to produce a corresponding quaternary carbonate, and
 - (b) a 2nd step of mixing the quaternary carbonate thus formed with an acid, while removing carbon dioxide generated from the system, to anion-exchange the carbonate into the corresponding acid salt.
2. The process as claimed in claim 1, wherein the acid which is mixed with the quaternary carbonate is an organic acid.

Revendications

1. Procédé pour la préparation d'un sel quaternaire comprenant
 - (a) une première étape de réaction d'une phosphine avec un diester de l'acide carbonique pour préparer un carbonate quaternaire correspondant, et
 - (b) une seconde étape de mélange du carbonate quaternaire ainsi formé avec un acide, pendant que l'on élimine du système le dioxyde de carbone, pour faire un échange d'anions du carbonate au sel d'acide correspondant.

2. Le procédé selon la revendication 1, dans lequel l'acide qui est mélangé avec le carbonate quaternaire est un acide organique.

Patentansprüche

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1. Verfahren zur Herstellung eines quartären Salzes mit folgenden Schritten:

(a) Reaktion eines Phosphins mit einem Carbonsäurediester in einem ersten Schritt zur Herstellung des korrespondierenden quartären Karbonates; und

10 (b) Mischen des so gebildeten quartären Karbonates mit einer Säure in einem zweiten Schritt, während das erzeugte Kohlendioxid aus dem System entfernt wird, um das Karbonat in das korrespondierende Säuresalz durch Anionenaustausch umzuwandeln.

2. Verfahren nach Anspruch 1, dadurch **gekennzeichnet**, dass die Säure, die mit dem quartären
15 Karbonat vermischt wird, eine organische Säure ist.

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